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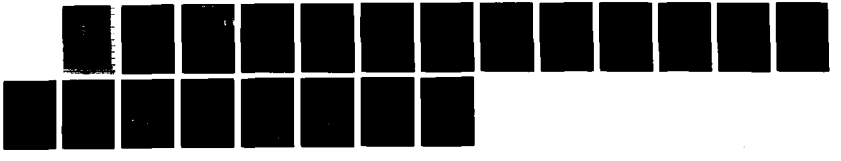

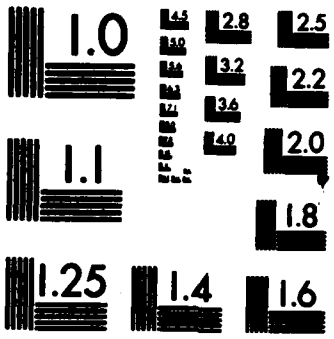

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Annual Report for contract N00014-86-K-0711

Ultrafast processes and spectroscopy with free electron lasers

prepared by

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The goal of this contract is to demonstrate that free-electron lasers (FELs) can be used and provide unique advantages for the study of ultrafast processes by time-resolved spectroscopy in the picosecond and even femtosecond time domain. The materials that will be investigated are amorphous semiconductors, including a-Si:H, a-(Si,Ge):H alloys and superlattices. These samples are grown in Professor S. Wagner's laboratory in the department of electrical engineering at Princeton, with whom we have been collaborating actively for

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Our goal is to demonstrate the power of FEL's in standard picosecond and femtosecond spectroscopy by performing time-resolved reflectivity or transmission, and wavemixing experiments in semiconductors. Of special interest to us are the carrier relaxation and trapping times in amorphous semiconductors.

We report progress on three fronts. First, we have established a permanent laboratory at Stanford University, where we have assembled the necessary equipment. Second, we have developed a novel method to measure accurately and over an extremely wide frequency range the duration and coherence properties of ultrashort laser pulses. This technique has been demonstrated and analyzed successfully at Princeton and appears ideally suited to the characterization of FEL's. Third, results from our recent femtosecond optical spectroscopy studies of amorphous silicon have helped us refine our experimental goals in this program, which will be discussed.

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the past three years.

In the first year of this contract, we have made progress on three fronts. First, we have established a permanent base at Stanford University where part of the equipment necessary for this project has been assembled. Second, we have developed a novel method for accurate determination of pulse duration and coherence time over a very wide frequency range, from the IR to the UV. Finally, we have performed picosecond and femtosecond time-resolved experiments with "conventional" lasers in materials of interest to this program.

Our FEL experiments will be performed on the SCA free electron laser at Stanford University. We have now an equipment base at Stanford where a large fraction the capital equipment and the supplies and expandables budgeted for the first year are presently located. We have also participated to three runs, all of which have unfortunately not produced useful light. The accumulated time spent by graduate students at Stanford is of the order of 6 weeks. As soon as the FEL is running again (around october 1st), we will be in position to start experiments immediately.

Because we want to demonstrate the usefulness of FELs as a tunable, high peak and average power ultrashort light pulse source, we

must first characterize fully the pulses. We need to know the pulse duration, the coherence time, the stability within a macropulse and from macropulse to macropulse. We have now demonstrated a novel technique to characterize the pulse duration and coherence time of ultrashort light pulses. This technique is called self-diffraction. Basically, the laser beam is split into two equal intensity arms which are made to intersect on an absorbing sample where they write a transient population or heat grating. A delay line introduces a variable time delay between the two writing pulses, which translates into a variable diffraction efficiency. The diffraction efficiency is measured either by a third pulse from the same laser or more conveniently by one of the two writing pulses. The diffracted energy as a function of time delay is made of a spike at zero delay and of a background which may or may not be centered at zero delay, depending on the configuration. The spike measures the coherence properties of the pulse and the background measures the pulse duration. Besides simplicity and sensitivity, this technique has the tremendous advantage of requiring *absorbing* media, not transparent crystals. It can thus be used everywhere in the spectrum, including in the UV and VUV. We plan to use this technique to characterize the FEL pulses at Stanford as soon as lasing is achieved. Our work has been presented as a poster paper at

the Conference on Lasers and Electro-Optics held in Baltimore in April 1987. We have almost finished writing a letter that includes some of our results, which we plan to send to Optics Communications within one month. In this work, we have been collaborating with Dr. Rick Trebino from Sandia National Labs in Livermore, who has supplied part of the theoretical analysis.

Under this contract, we have also performed two separate time-resolved experiments. In one experiment, we have for the first time measured the complex dielectric function of molten silicon. High power picosecond pulses from a Nd:YAG laser produced a short-lived liquid layer on the surface of a wafer. The reflectivity of a time-delayed probe pulse at the fundamental, second or third harmonic of the YAG was measured as function of angle of incidence. In this way, both the real part and the imaginary part of the dielectric function (or index of refraction) can be obtained. The relaxation time and the plasma frequency of liquid silicon is then measured. In particular, the relaxation time is approximately 0.2 femtoseconds. We have briefly presented some of these results at the International Quantum Electronics Conference held in Baltimore in April 1987. We have just finished writing a letter summarizing our results, which has been sent to Applied Physics Letters. A copy of this letter is attached to this

annual report. In the second experiment, we have used the tools of femtosecond time-resolved spectroscopy to study some ultrafast electronic processes in a-Si:H. Without support from this contract, we had published our first results in a Physical Review Letters late in 1986. Although we now have more results that confirm our original model and increase our understanding of carrier thermalization in the extended states and of trapping in the weakly localized states, the unavailability of ultrashort IR (wavelength longer than 1 micron here) pulses makes our interpretation more difficult and prevents us from accessing part of the physics. This is one of the motivations for using an FEL and we hope to remedy that problem during the next year when the SCA FEL is operational. Our latest results have also been presented at the International Quantum Electronics Conference in Baltimore.

The Drude parameters of liquid silicon at melting temperature

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ABSTRACT

The Drude parameters of liquid silicon at melting temperature have been obtained from time-resolved reflectivity measurements at 1064, 532 and 355 nm following melting of an optically thick layer by a picosecond visible laser pulse. The ratio of the electron density N to the electron mass m is found to be equal to 2.17 $10^{50} m^{-3} kg^{-1}$ and the relaxation time τ to be equal to 212 attoseconds. These values confirm previous results obtained by cw ellipsometry between 1 and 0.4 microns.

In contrast to solid silicon, which is a semiconductor, liquid silicon is a metal, characterized by a high reflectivity at normal incidence around the visible. This fact is now widely known thanks to the large body of work done recently on pulsed laser annealing. However, liquid silicon had been studied for some time and some of its properties can be found in reference 1. The melting temperature of silicon is 1683 K, which unfortunately creates serious experimental problems for an accurate measurement of any parameter.

The plasma frequency and the relaxation time are the two basic parameters describing the properties of any Drude metal. They are related to the complex dielectric function (or index of refraction) through the well-known Drude formulae. Shvarev at al. [2] have reported ellipsometric measurements of ω_p and τ from 1 micron to 0.4 micron. Their measurements were performed on a molten pool of silicon held approximately at melting temperature. Their data could be fitted with a simple Drude model, where $\omega_p = 2.56$ 10^{16} rad/s and $\tau = 217$ as (1 as $= 10^{-18}$s). However, they did not assign error bars to their determination. Recently, Jellison and Lowndes [3], and Gusakov et al. [4] have performed nanosecond time-resolved ellipsometric measurement at 632.8 nm during nanosecond pulsed laser melting of silicon. Their results were in fair agreement with those of Shvarev. We have also recently reported picosecond time-resolved reflectivity measurements with *p*-polarized 1064 nm pulses upon melting of silicon with a picosecond laser beam [5]. Our results were in good agreement with those of Shvarev et al.

In this letter, we report our determination of the plasma frequency ω_p and relaxation time τ of molten silicon at the melting temperature. We have measured the reflectivity of silicon after melting with a picosecond laser pulse at 532 nm. Three wavelengths were used, 1064, 532 and 355 nm corresponding to the fundamental, second and third harmonic of our Nd:YAG laser. In each case, the measurement was repeated

at different angles of incidence with a *p*-polarized probe beam. To avoid problems due to overheating of the liquid phase, the energy density of the melting beam was kept close to the melting threshold. Even several hundred picoseconds after melting, when an optically thick liquid silicon layer exists at the melting temperature, we are not concerned by the presence of an oxide layer, by the presence of vapor, or by significant surface bulging.

Our experimental method is as follows. Thirty to sixty picosecond long pulses are produced by an actively-passively modelocked Nd:YAG laser followed by a double pass amplifier. The pump pulse, which melts the wafer, is obtained after frequency doubling in a KDP crystal. The probe pulse, whose reflectivity is measured, is either at the fundamental, second or third harmonic frequency of the laser. The pump pulse is near normal incident on the sample whereas the angle of incidence of the *p*-polarized probe pulse is varied from near normal incidence to near grazing incidence. Time resolution is achieved by delaying the pump pulse with a computer-controlled delay line. The reflectivity of the probe pulse is measured with a *pin* silicon photodiode and read off a sample and hold circuit by the computer. The sample is moved after each shot to provide a fresh spot. The samples are polished silicon wafers with low to moderate doping. The native oxide layer has not been removed.

We want to measure two quantities, the plasma frequency ω_p and the relaxation time τ. These two quantities can be determined directly from the knowledge of the real and imaginary parts of the dielectric function by the well-known Drude formulae

$$\epsilon_R = 1 - \frac{\omega_p^2 \tau^2}{1+\omega^2\tau^2} \tag{1}$$

$$\epsilon_I = \frac{\omega_p^2 \tau}{\omega(1+\omega^2\tau^2)} \tag{2}$$

$$\omega_p^2 = \frac{Ne^2}{m\epsilon_0} \tag{3}$$

where N is the electron density, m is the effective electron mass, e is the electron charge ($1.6\ 10^{-19}$ C), ϵ_0 is the dielectric constant of vacuum ($8.854\ 10^{-12}$ F/m) and ω is the probe laser frequency. To determine both ϵ_R and ϵ_I, we must make at least two measurements. Ellipsometry is one possible method, since it provides information about the amplitude and phase of the reflected beam. Ellipsometry is best suited to cw measurements, although it has been demonstrated with nanosecond [3,4] and even picosecond [6] pulses. An easily implemented alternative technique is to measure the reflectivity at several (at least two) angles of incidence [5]. In order to get the largest contrast between the reflectivity in the solid and liquid phases (and hence the most accurate determination of the ratio), we use *p*-polarized light and perform the experiment with at least several angles close to Brewster's angle for the solid [7].

In Figure 1, we show typical results obtained with each probe frequency in the neighborhood of Brewster's angle. The measurements were obtained after melting with a pump pulse slightly exceeding melting threshold. The full line is calculated from the reflectivity in the solid phase using the data of reference 8 and in the liquid phase using the data of reference 2 and accounts for the finite probe duration. The plateau in the reflectivity which is observed 100 ps or so after pumping is taken to be the signature of an optically thick liquid layer at or very close to the melting temperature. This is in agreement with one dimensional heat flow calculations that we have performed. Each data point is the average of three or more measurements and the error bar represents the standard deviation. To obtain an accurate value for the reflectivity of the liquid phase, we compute the average value in the reflectivity plateau. When we do so, we obtain the results of Figure 2. In that figure, we show the measured reflectivity as a function of angle for the three probe wavelengths, together with the calculated reflectivity in the solid phase [8] and in the liquid phase [2].

From Figure 2, we can immediately observe the good qualitative agreement between our measurements and the Drude constants of reference 2. In order to find the best values for ω_p and τ, and to evaluate the accuracy of our determination, we first find the region in the (ϵ_R, ϵ_I) plane which leads to calculated curves falling within the error bars of our measurements. This region maps into another region in the (ω_p, τ) plane. We then repeat the procedure for each wavelength. Assuming that liquid silicon at melting temperature is indeed a Drude metal, ω_p and τ are wavelength-independent. The three regions intersect in the (ω_p, τ) plane and we interpret this intersection as defining the range of values for ω_p and τ which are acceptable. Finally, we substitute for the plasma frequency the ratio of the electron density to the electron mass, which are related through equation 3. Our results are compared to the results of Shvarev et al. in Table 1. In the (N/m, τ) plane, the acceptable values span a triangular region defined by (224.8, 202), (219.6, 214), (207.5, 220), where N/m is in units of $10^{57} m^{-3} kg^{-1}$, and τ is in attoseconds (1 as = 10^{-18} s). The values listed in Table 1 correspond to the center of gravity of that triangular region.

Since Shvarev et al. did not specify the uncertainty in their measurements, we can consider our results and their results to be in perfect agreement. In particular, at 632.8 nm, we calculate ϵ_R = -19.2 and ϵ_I = 32.0, in much better agreement with Shvarev's values of -20.8 and 33.7 than with the nanosecond time-resolved ellipsometeric data of Jellison and Lowndes of -12 and 40. Note that similar nanosecond time-resolved ellipsometric measurements by Gusakov et al. who found ϵ_R = -21.2 and ϵ_I = 30.5 at 632.8 nm also agree well with Shvarev's data and our results. Finally, since for each probe wavelength, we are either in the classical skin effect region (at 1064 and 532 nm) or in the relaxation region (at 355 nm) [9], our analysis is correct as it stands.

In conclusion, we have measured the Drude parameters of liquid silicon at or close to the melting temperature using the methods of picosecond time-resolved spectroscopy, in the near infrared, in the visible and in the near ultraviolet. We find that the ratio of the electron density N to the electron mass m is equal to 217 $10^{57} m^{-3} kg^{-1}$ and that the relaxation time τ is equal to 212 attoseconds. Each value is accurate within $\pm$ 5%. Our results indicate the absence of gaps or other features in the band structure of liquid silicon, even in the near ultraviolet. This research was supported in part by an IBM Faculty Development Award with additional support from ONR contract N00014-86-K-0711. We thank W.L. Nighan, Jr. and N.K. Bambha for technical assistance.

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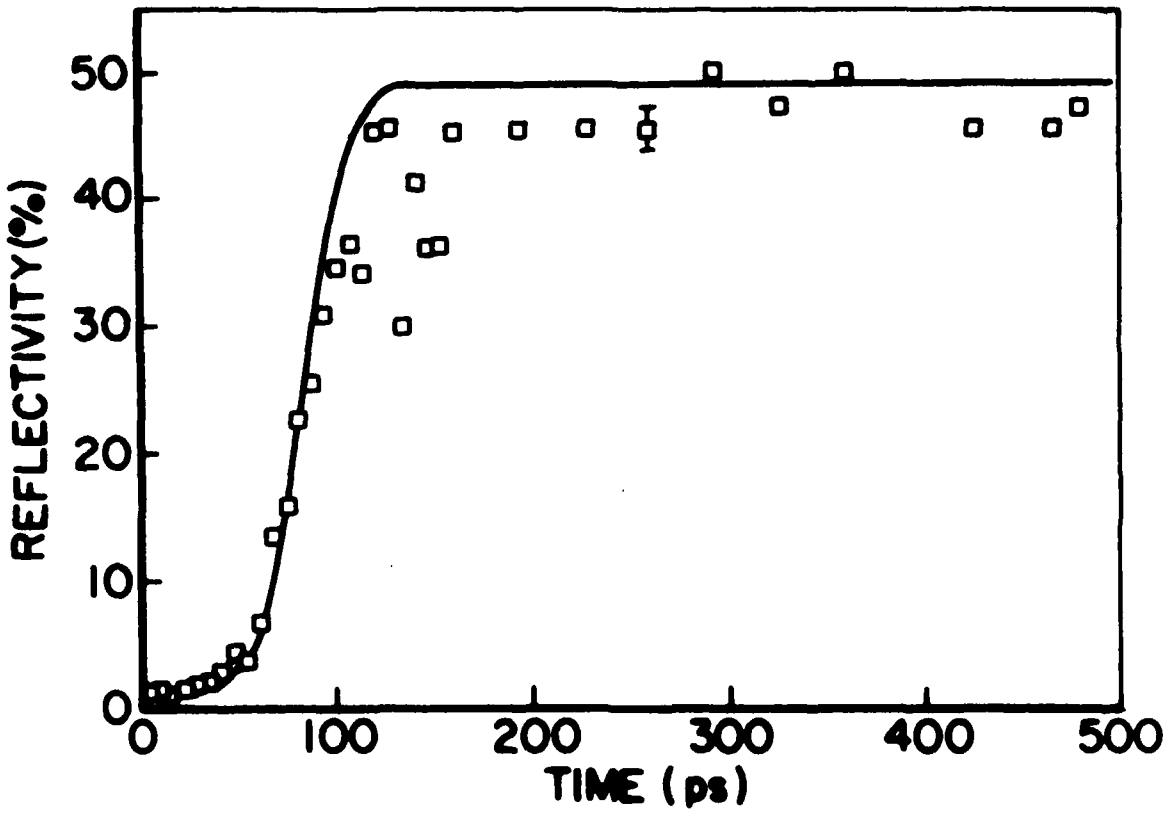
Figure Captions

Figure 1: Time-resolved reflectivity measurements after melting with a 532 nm pump pulse. The experimental conditions are (a) λ_p = 1064 nm, $\theta = 70^\circ$; (b) λ_p = 532 nm, $\theta = 70^\circ$; (c) λ_p = 355 nm, $\theta = 63^\circ$.

Figure 2: Reflectivity of molten and solid silicon as a function of angle of incidence θ for *p*-polarized light at 1064 nm, 532 nm and 355 nm. The full curve is calculated from the data of Shvarev et al (Ref 2) and the dashed curve from the tabulated data in Ref 8. The experimental points and error bars are obtained by the average and standard deviation of the reflectivity plateau. The error bar for the angle is a conservative estimate based on the angular spread of the strongly focussed probe beam and the accuracy in determining the angle.

Table Caption

Table 1: Comparison between our data and Shvarev's determination (Ref 2) of ω_p and τ.


REFLECTIVITY(%)
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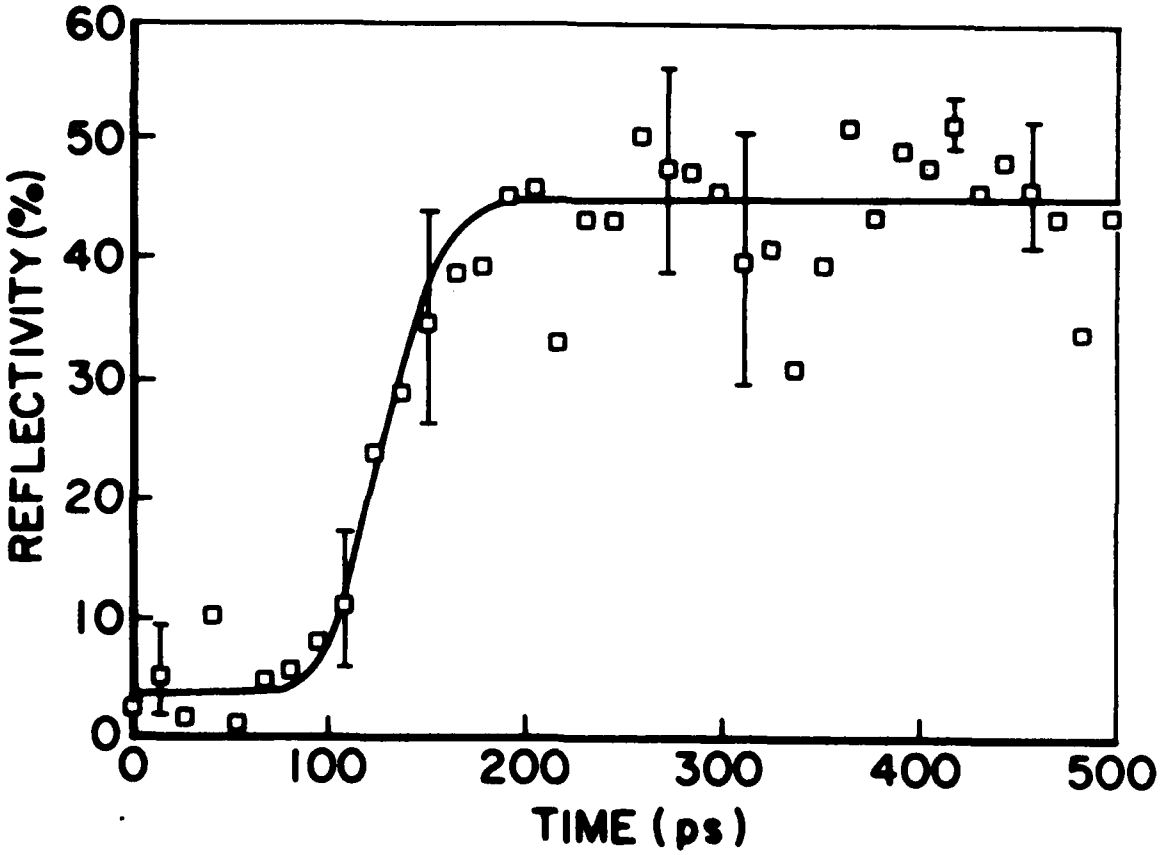
40
30
20
10
0
0
100
200
300
400
500
TIME (ps)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50
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30
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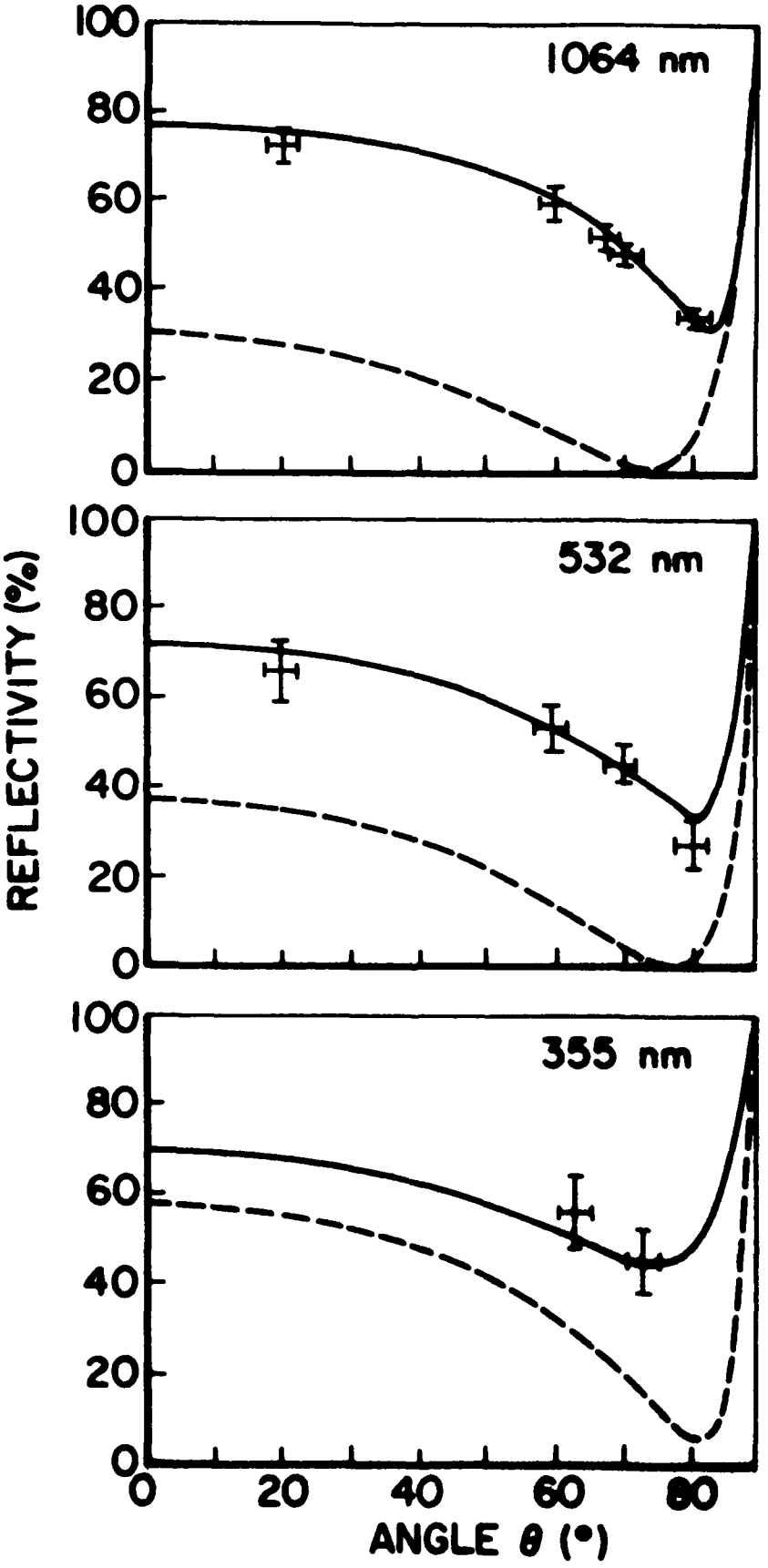
10
0
REFLECTIVITY (%)
0
100
200
300
400
500
TIME (ps)


1064 nm
532 nm
355 nm
REFLECTIVITY (%)
ANGLE θ (°)
0
20
40
60
80
100

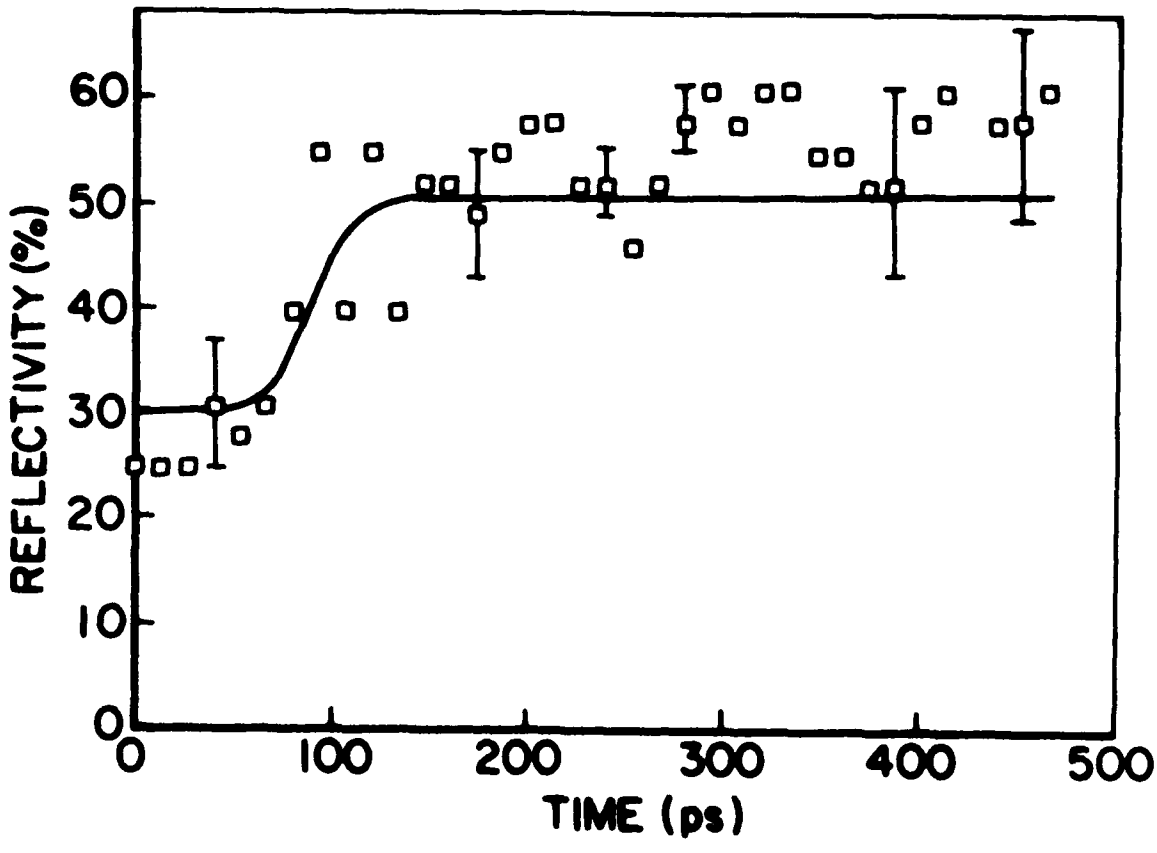

REFLECTIVITY (%)
60
50
40
30
20
10
0
0
100
200
300
400
500
TIME (ps)

Table 1

	Shvarev et al (Ref 2)	This Work
$\omega_p(10^{16}\text{rad/s})$	2.56	$2.50 \pm 5\%$
$\tau\,(10^{-18}\text{s})$	217	$212 \pm 5\%$

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